Photonic Crystal Surface Emitting Lasers across the Visible Spectrum based on Colloidal Quantum Dots

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***Abstract*— Combining integrated optical platforms with solution processable semiconductor materials offers a clear path towards miniaturized and robust light sources, including lasers. Semiconducting colloidal quantum dots present a unique platform to realize this by combining tunable properties, high luminescence efficiency. A limiting aspect for both red and green emitting materials remains the drop in efficiency at high excitation density due to non-radiative quenching pathways, such as Auger recombination. Next to this, lasers based on such materials remain ill characterized, leaving questions on their ultimate performance. Here, we show that weakly confined 'bulk' colloidal quantum dots offer a unique solution processable materials platform to circumvent the long-standing material issues. Here, we first show that optical gain in such systems is mediated by a 3D plasma state of unbound electron-hole pairs which gives rise to broadband and sizable gain across the full red spectrum with record gain lifetimes and low threshold. As proof of concept, the nanocrystals are integrated on a silicon nitride platform enabling high spectral contrast, surface emitting and TE polarized PCSEL – type lasers with ultra-narrow beam divergence across the visible (green, red) spectrum from a small surface area. Our results prime QS materials as excellent materials platform to realize highly performant and compact on-chip light sources.**

Keywords — PCSEL, nanocrystals, confinement, solution processable, green lasers, red lasers.

I. Introduction

Optoelectronics based on solution-processable materials, such as small-molecule organics or strongly confined colloidal quantum dots (QDs) – a recent Nobel Prize in Chemistry winning class of low dimensional semiconductor nanocrystallites - holds great promise for low-cost, flexible optoelectronic devices. Over the past 20 years, such promise was indeed met in the fields of LEDs and photo-detection where light-matter interaction is dictated by linear absorption and spontaneous emission. Pushing these materials to a net optical gain remains, however, a huge challenge as of today. Inorganic nanocrystals are probably the most promising candidates to achieve stable room-temperature lasing due to a unique combination of a long-term structural stability and an electronic structure tunable by size, shape and composition. After two decades of research several promising results indicate a way forward, such as electrically pulsed amplification. However, a main hurdle stone remains the fast non-radiative recombination at high carrier density, mainly Auger limited, combined with limited (modal) gain coefficients – a bottleneck in thin film technologies where electrode losses overshadow material gain. Colloidal 'bulk' nanocrystals – nanocrystals with dimensions larger than the Bohr diameter and hence not subject to strong confinement - might offer a way out of this predicament by providing high material gain, translating into high modal gains, combined with suppressed non-radiative recombination.

II. Results & Discussion

To cover the green spectrum, colloidal 'bulk' nanocrystals (BNCs) of CdS were synthesized in-house, see Figure 1a for a TEM micrograph. For the red part of the spectrum, we employ CdSe nanocrystals. Both are 'bulk' in the sense that their optical properties don't depend on their size anymore, opposing the wealth of reports on (strongly) confined alternatives. In practice this boils down to making the nanocrystals with sizes above 10 nm. Both systems are characterized using quantitative pump-probe spectroscopy, as shown in Figure 2.

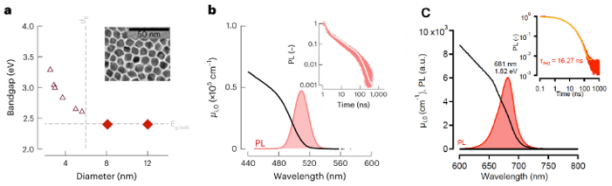

Figure 1: *(a) Sizing curve showing band gap (in eV) versus particle diameter of CdS. Red markers are the samples used which are in the regime of vanishing carrier confinement. (b) absorption and photoluminescence of a dispersion of CdS BNCs. Inset shows their luminescence decay profile after 400 nm excitation.(c) similar data for the CdSe BNCs active in the red spectrum.*

The data shown in Figure 2 indicates that a material gain, normalized for volume fraction and assuming 100% mode confinement, can reach values up to 50.000 cm^{-1} . The net material gain can be maintained up to 3 ns, a record for colloidal nanomaterials. Similar results are obtained for CdSe (not shown) albeit with smaller material gain coefficients.

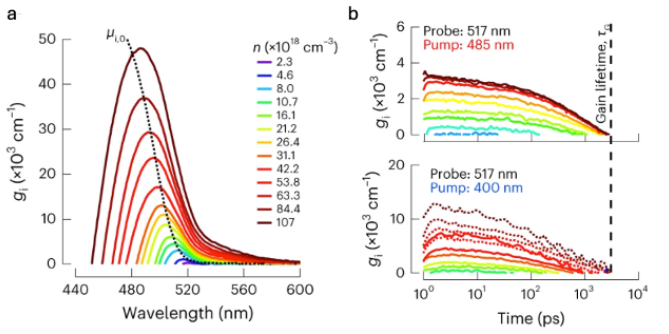

Figure 2: *Pump Probe spectroscopy measuring normalized absorbance to represent the material gain g_i. (a) Spectra at 3 ps delay and (b) kinetics at 517 nm after femtosecond 400 nm pumping.*

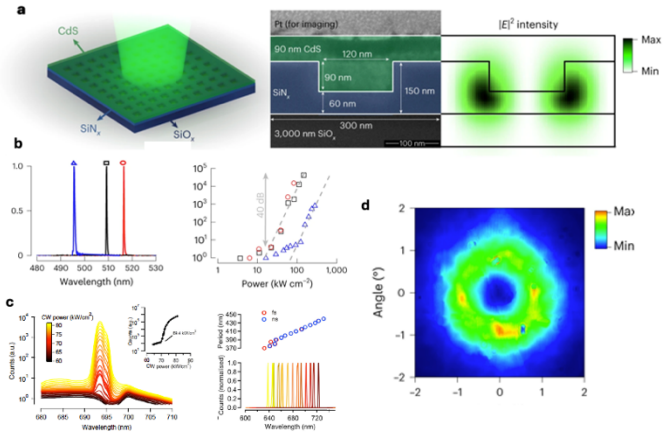

Figure 3*: PCSEL lasers based on BNC nanocrystals. (a) 2D grating etched into silicon-nitride with CdS BNC layer on top. ght shows a SEM cross section and FDTD simulation of the electric field intensity. (b) lasing spectra collected normal to the surface for varying pitch of the grating with output versus pump power (in kW/cm²) for excitation with 7 ns pulses at 355 nm. (c) Similar data for CdSe red emitting PCSEL architecture with inset showing pump power dependence. Right shows the variation of lasing wavelength with pitch of the grating. (d) Far field profile translated into divergence angle.*

In a next step, we combined these efficient optical gain materials with an integrated photonics platform, i.e. silicon nitride based micro-chips. By etching a periodic grating into the nitride surface and overcoating it with a simple spin coating from the BNC colloidal dispersions , very much like a simple ink deposition, we can obtain optically pumped lasing in the green and red with ca. 10 kW/cm^2 thresholds from sub 100 square micrometer areas. The lasers show a high dynamic range and single mode behavior combined with a highly collimated light output in the notorious green gap (485 – 520 nm) and in the red (640 – 720 nm).

III. Conclusion

Our results prime weakly confined nanocrystals – so called ‘bulk nanocrystals’ as excellent optical gain materials to enhance integrated photonic platforms such as silicon nitride. After 20 years of research into the optimal nanoscale architecture for lasing for solution processable nanocrystals, our work shows that actually reducing confinement is the best way to go. Downsides of the ‘bulk’ route are a lack of tunability through size variation, a point remedied by varying the composition of the material (e.g. CdS and CdSe or by alloying).

References

[1]Tanghe, I., Samoli, M., Wagner, I. et al. *Optical gain and lasing from bulk cadmium sulfide nanocrystals through bandgap renormalization*. Nature Nanotechnology **18**, 1423–1429 (2023).

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